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Spectral Hole-Burning of Eu^{3+} : Y_2SiO_5 Crystal at 579.62 nm*

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Abstract: By using an Ar^+ ion laser, a tunable Rh 6G dye laser (Linewidth: 0.5 cm^{-1}) and a Coherent 899-21 dye laser as light sources and using a monochromator and a phase-locking amplifier, the optical properties of Eu^{3+} : Y_2SiO_5 crystal were detected. Persistent spectral hole burning (PSHB) were also observed in $^5D_0$-$^7F_0$ transition in the crystal at the temperature of 16 K. For 15 mW dye laser (Wavelength: 579.62 nm) burning the crystal for 0.1 s a spectral hole with about 80 MHz hole width were detected and the hole can been keep for longer than 10 h.

Key words: Eu^{3+} ion : Y_2SiO_5; spectral hole burning (PSHB)
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Trivalent europium ions doped in crystals are often used for the study of high-resolution nonlinear spectroscopy because Eu^{3+} ion doped in crystals have the longest optical dephasing times t_2. A material with long dephasing time t_2 can be used for frequency-selective optical memory, both in the frequency domain (persistent spectral hole burning) and the time domain (photon echo memory)[1]. The value of t_2 of impurity ions in solids mainly depends on (1) guest-ion/phonon interaction, (2) guest-ion/guest-ion interaction, and (3) guest-ion/host-ion interaction. Because the effect of phonons becomes smaller at low temperature the value of t_2 can becomes temperature independent by lowering temperature. The mutual interaction between guest ions can be decreased by doping crystals with low rare earth ion concentrations. The interaction between guest ions and host ions may be reduced by choosing appropriate host crystals composed of elements with small nuclear moment. Eu^{3+} : Y_2SiO_5 crystal is a newly luminescence material. Because the nuclear moments of the composite elements are small in this crystal, the spin-spin interaction between the guest ion and the host ion is minimized. The guest-host interaction does not cause large additional line broadening, therefore this crystal is good optical data storage material. The spectra and spectral hole-burning of Eu^{3+} : Y_2SiO_5 crystal were investigated in this paper.

1 Experimental Results

The aim of this work was to study the spectra and the spectral hole burning of Eu^{3+} : Y_2SiO_5 crystal. Eu^{3+} ions-doped concentration was 0.15% in our sample. The emission spectra of the Eu^{3+}-doped Y_2SiO_5 crystal from 560 to 720 nm were measured using a Coherent Innova 10 argon ion laser (Line 514.5 nm) excitation. The luminescence was dispersed by a Jobin-Yvon HR-1500 model monochromator and detected by a SR565 phase-locking amplifier.

Fig. 1 shows the fluorescence spectra of $^5D_0 \rightarrow {}^7F_{0,1,2}$ transitions. Fig. 2 shows the fluorescence spectra of $^5D_0 \rightarrow {}^7F_{3,4}$ transitions.

The photoluminescence excitation spectra of $^7F_0 \rightarrow {}^5D_0$ transition excited by a Q-switched Nd^{3+} : YAG laser pumped tunable Rhodamine 6G dye laser with 0.5 cm^{-1} linewidth were studied by receiving fluorescence of $^5D_0 \rightarrow {}^7F_2$ transition whose central wavelength is about 615 nm. Eu^{3+} ions occupy two optical sites whose spectral lines of $^7F_0 \leftrightarrow {}^5D_0$ transition are about 0.2 nm away from each other at room temperature and both sites are the luminescence centers with C_1 symmetry. The site selective fluorescence spectra of Eu^{3+} ion $^5D_0 \rightarrow {}^7F_0$ transition were detected by exciting each site respectively.

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Hole burning experiments were done on the excitation spectrum of the $^7F_0$-$^5D_0$ transition of the Eu^{3+} ion. Low temperature persistent spectral hole burning was observed by irradiating the crystal at 16 K by a tunable cw Ar^+-ion-laser-pumped Rhodamine 6G dye laser (Coherent 899-21 laser) with 1 MHz linewidth.

A tunable Rh 6G dye laser with 0.5 cm^{-1} spectral linewidth pumped by the second harmonic of a pulsed Nd^{3+} :YAG laser was used as the excitation light source, a boxcar and a computer were used as data detecting system. Photoluminescence excitation spectrum of $^5D_0 \rightarrow {}^7F_0$ of the crystal were detected. It is plotted in Fig. 3. Two spectral peaks (Site 1: 579.62 nm and site 2: 579.82 nm) that suggest that Eu^{3+} ion occupies two Y^{3+} ions site in the Y_2SiO_5 crystal are shown in the excitation spectrum. Site selective fluorescence spectra were also detected in our experiment. They were displayed in Figs. 4(a) and (b). Fluorescence spectra of $^5D_0 \rightarrow {}^7F_{1,2,3,4}$ transitions can clearly be seen in the site selective fluorescence spectra . By studying the selective fluorescence spectra we found that the positions of spectral peaks in the Fig. 4(a) were different from them in Fig. 4(b). The fluorescence spectra of different sites does not overlap each other.

Y_2SiO_5 belongs to the monoclinic biaxial crystal class 2/m (Space group $C2/c$)[2]. The lattice parameters were given in Refs. [3,4]. Tkachuk et al[5] report that Nd^{3+} ions occupy two distinct optical sites; they have also made resolved Stark level measurements on both sites. Because of the biaxial character of the system three principal axes of polarization can be defined: (1) the <010> direction which is a two fold symmetry axis of the crystal and (2) the D1 and D2 directions perpendicular to each other and to the <010> direction and which correspond to extinction directions[4] when the sample is viewed in the <010> direction between crossed polarizers. These directions were determined exactly with respect to the crystallographic axis by X-ray analysis and shown in Ref. [4].

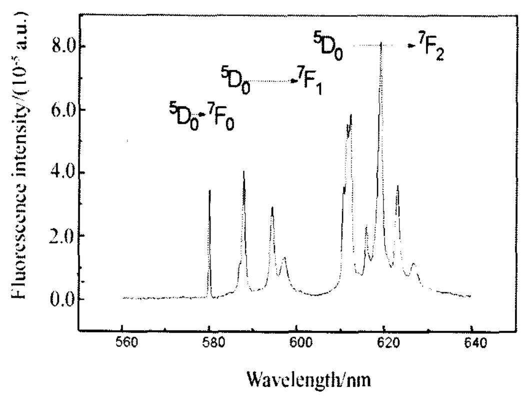

Fig. 1 Fluorescence spectra of $^5D_0 \rightarrow {}^7F_{0,1,2}$

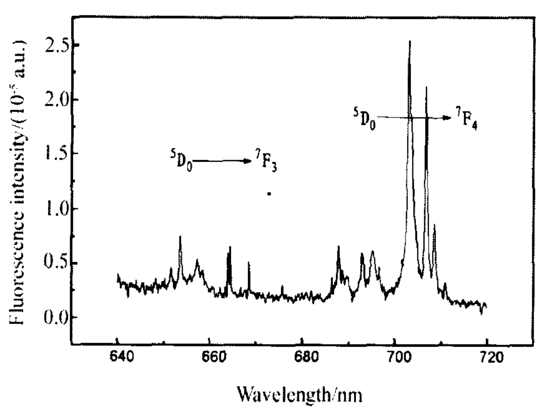

Fig. 2 Fluorescence spectra of $^5D_0 \rightarrow {}^7F_{3,4}$

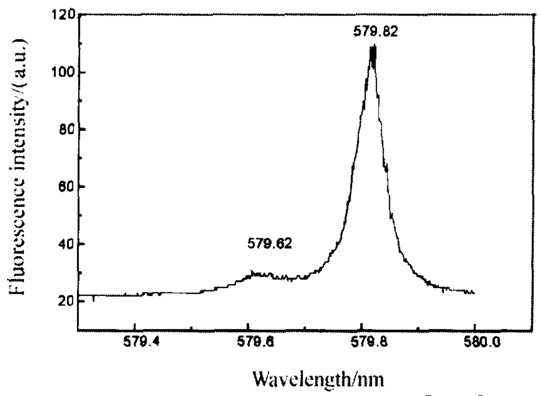

Fig. 3 Photoluminescence excitation spectra of $^7F_0 \rightarrow {}^5D_0$ transition of Eu^{3+} :Y_2SiO_5 crystal (Em: 615 nm)

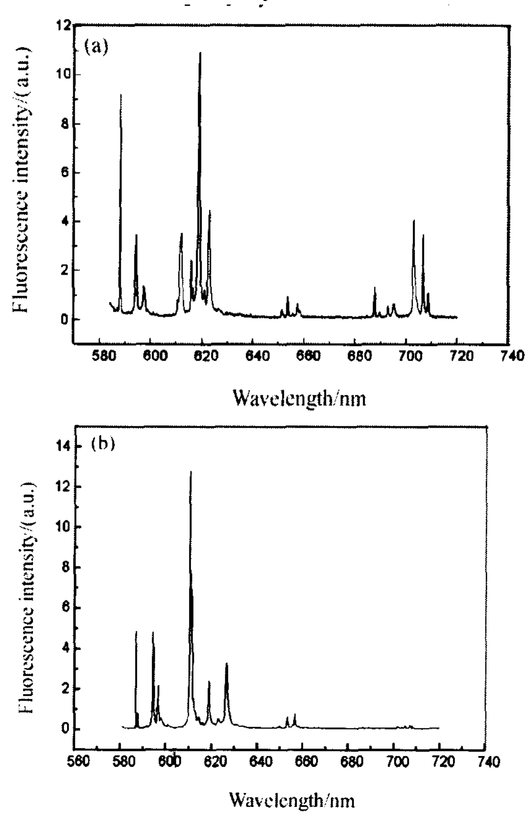

Fig. 4 Site selective fluorescence spectra of site 1(a) (Ex: 579.62 nm) and 2 (b) at temperature of 77 K (Ex: 579.82 nm)

Fig. 5 shows that the XRD patterns of the crystal which contain more than ninety diffraction peaks. Table 1 displays the lattice constants *a*, *b*, *c* and β calculated by formula

$$\frac{1}{d^2}=\frac{1}{\sin^2\beta}\left(\frac{h^2}{a^2}+\frac{k^2\sin^2\beta}{b^2}+\frac{l^2}{c^2}-\frac{2hl\cos\beta}{ac}\right)$$

where *d* is the interplanar distance and *h*, *k*, *l* are lattice indexes.

A Coherent 899-21 dye laser with 1 MHz spectral linewidth pumped by a Ar^+ ion laser was used for spectral hole-burning of $Eu^{3+}:Y_2SiO_5$ crystal. The system of a Jobin-Yvon HR-1500 model monochromator, a SR565 phase-locking amplifier and a computer was used for receiving and printing transmission spectra.

Fig. 6 shows the experiment setup of spectral hole burning. A spectral hole with about 80 MHz hole width are observed after 15 mW dye laser (Wavelength: 579.62 nm) with 1 MHz linewidth burns $Eu^{3+}:Y_2SiO_5$ crystal for 0.1 s at the temperature of 16 K.

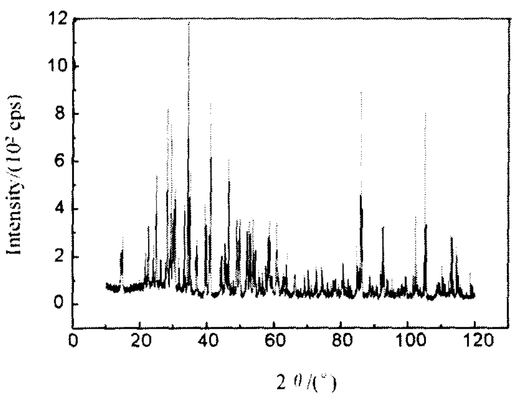

Fig. 5 Pattern of X-ray diffraction of $Eu^{3+}:Y_2SiO_5$ crystal

Table 1 Crystal lattice constants of $Eu^{3+}:Y_2SiO_5$

Parameters	Doped	Undoped *
a/nm	1.2533 (85)	1.25013 (17)
b/nm	0.6735 (36)	0.67282 (8)
c/nm	1.0435 (80)	1.04217 (13)
β/(°)	102.58 (72)	102.682 (11)

* Taken from Ref. [4]

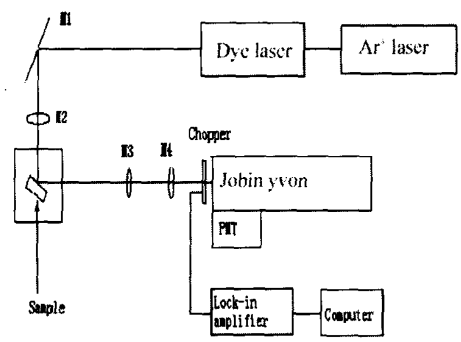

Fig. 6 Experiment setup of spectral hole burning

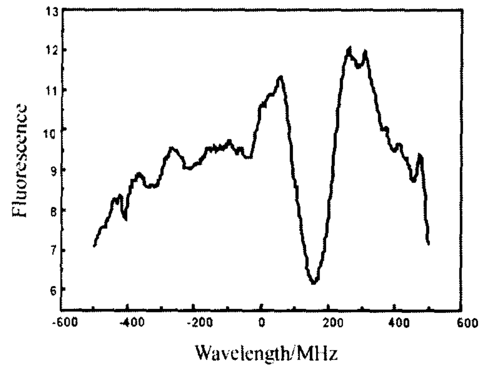

Fig. 7 Single hole in absorption line of $^5D_0$-$^7F_0$ transition of site 1 at temperature of 16 K (Linewidth of scanning dye laser: 1 MHz, hole burning laser wavelength: 579.62 nm, the power of hole-burning laser: P_h = 15 mW, the power of scanning laser: P_d = 0.1 mW, the receiving wavelength of monochromator: λ_{em} = 615 nm)

Hole spectra were recorded by scanning the dye laser near 579.62 nm while monitoring the fluorescence of $^5D_0 \rightarrow {}^7F_2$ transition at 615 nm, as shown in Fig. 7. The laser power for scanning was 0.1 mW. The spectral hole can last for longer than 10 h at the temperature of 16 K.

2 Conclusion

Two absorption peaks in photoluminescence excitation spectrum of $^5D_0 \rightarrow {}^7F_0$ transition show that Eu^{3+} ion occupies two Y^{3+} ions sites in the Y_2SiO_5 crystal. Spectral hole is burnt at higher temperature (16 K) while it is usually burnt at 4.3 K[1] in $Eu^{3+}:Y_2SiO_5$ crystal. Spectral hole can keep for longer than 10 h in $Eu^{3+}:Y_2SiO_5$ crystal at 16 K.

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